

# Investigation of the fundamental constants stability based on the reactor Oklo burn-up analysis

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The burn-up for SC56-1472 sample of the natural Oklo reactor zone 3 was calculated using the modern Monte Carlo codes. We reconstructed the neutron spectrum in the core by means of the isotope ratios:  $^{147}\text{Sm}/^{148}\text{Sm}$  and  $^{176}\text{Lu}/^{175}\text{Lu}$ . These ratios unambiguously determine the spectrum index and core temperature. The effective neutron absorption cross section of  $^{149}\text{Sm}$  calculated using this spectrum was compared with experimental one. The disagreement between these two values allows to limit a possible shift of the low laying resonance of  $^{149}\text{Sm}$  even more. Then, these limits were converted to the limits for the change of the fine structure constant  $\alpha$ . We found that for the rate of  $\alpha$  change the inequality  $|\delta\dot{\alpha}/\alpha| \leq 5 \cdot 10^{-18}$  is fulfilled, which is of the next higher order than our previous limit.

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## I. INTRODUCTION

The confirmation of the temporal variation of the fundamental constants would be the first indication of the universe expansion influence on the micro physics. One of the possibilities is dependence of the nuclear couplings on the value of the cosmon field - a possible candidate for the dark energy (see [1, 2] and [3] for review) - the fundamental scalar field of high amplitude but tiny mass. If the equation of this field state  $w = p/\rho$  differs from -1 then the phenomenology of this field will be different from the universe with the cosmological constant. Then, the temporal dynamics of the state equation could be determined by investigation of the variation of fundamental constants with the increased accuracy [4]. The investigation of the natural reactor Oklo allow to bound the possible variation of the fine structure constant  $\alpha$  with record high accuracy [5, 6] and the deviation of  $w$  from -1 now ( $z \approx 0.13$ ) [7].

Shlyakhter was the first who showed that the variation of the fundamental constants could lead to measurable consequences on the Sm isotops concentrations in the ancient reactor waste [8]. Later Damur and Dyson [9] for Zones 2 and 5 and also Fujii [10] for Zone 10 of reactor Oklo made more realistic analysis of the possible shift of fundamental constants during the last  $2 \cdot 10^9$  years based on the isotope concentrations in the rock samples of Oklo core. In this investigation the idealized Maxwell spectrum of neutrons in the core was used. The efforts to take into account more realistic spectrum of neutrons in the core were made in works [11–13].

This paper continues the neutronics analysis of the Oklo reactor that we began in the previous work [12]. In paper [12] we considered a fresh core only and did not analyze the method of extraction of  $^{149}\text{Sm}$  absorption cross section by the geochemical analysis. Meanwhile, this cross section depends on the reactor burn-up and on the neutron spectrum transformation during the reactor operation. The bounds of the  $^{149}\text{Sm}$  resonance shift during the past  $2 \cdot 10^9$  years have been obtained here in a different way as compared to [12]. We used a modern reactor code to investigate the burn-up of a particular sample from Zone 3 of reactor Oklo. The conditions of the reactor operation could be determined according to the distribution of different nuclide in the sample. This procedure definitely determined the reactor spectrum based on which the theoretical effective neutron absorption cross section of the  $^{149}\text{Sm}$  nuclei can be calculated. The obtained effective cross section have considerably narrow uncertainties as compared to [12] where we vary the uranium content, water fraction and temperature of the reactor in the wide range. On the other hand, this cross section can also be evaluated by the isotopic ratios of the Samarium nuclide in the sample. Performing the fuel realistic burn-up we can determine the cross section of  $^{149}\text{Sm}$  needed to describe the concentration of this isotope in the sample. Comparing this value with the theoretical one we can make a conclusion about the variation of the  $^{149}\text{Sm}$  absorption cross section during the time after the Oklo phenomenon occurred.

A similar approach was used in [13]. However, in this paper the close limits on the temperature of the reactor core were accepted. To our opinion, this point needs the further investigation. Contrary, in our analysis the temperature of the reactor core during its operation is determined based on the lutetium isotope ratio. Also, the water content

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in [13] was not controlled. The uncertainties in the water concentration in the sample also considerably influence the result.

## II. DETERMINATION OF $^{149}\text{Sm}$ EFFECTIVE NEUTRON ABSORPTION CROSS SECTION

We use the following definition of the effective cross section [9]

$$\hat{\sigma} = \frac{\int \sigma(E)\Phi(E)dE}{v_0 \int n(E)dE}, \quad (1)$$

where  $\Phi(E)$  - flux of neutrons with energy  $E$ , and  $n(E) = \Phi(E)/v$  - neutron density at energy  $E$ ,  $\sigma(E)$  - the  $^{149}\text{Sm}$  nuclei absorption cross section of neutron. A neutron with energy 0.0253 eV has velocity  $v_0 = 2200$  m/sec. The integral in (1) is over the entire interval of neutron energies. Equation (1) determine the local instant definition of the effective cross section because the neutron flux  $\Phi(E)$  is different for the reactor core different areas and for different periods of the reactor operation. As was claimed earlier, this cross section strongly depends on the core temperature [9–12] and its element composition [11, 12].

Neutron absorption cross section of  $^{149}\text{Sm}$  at low neutron energies is saturated by the capture cross section. We use two resonance approximations of the neutron capture cross section for  $^{149}\text{Sm}$  by taken into account only the resonances at 97.3 meV and 0.87 eV energies. The Breit-Wigner formula of the capture cross section for one resonance as follows:

$$\sigma_{(n,\gamma)}(E) = g_0 \frac{\pi \hbar^2}{2mE} \frac{\Gamma_n(E) \cdot \Gamma_\gamma}{(E - E_\gamma)^2 + \Gamma_{tot}^2/4} \quad (2)$$

where  $g_0$  is the statistical factor,  $\Gamma_n, \Gamma_\gamma, \Gamma_{tot}$  - neutron widths (see [12] for resonance parameters). The calculated cross section is compared with the evaluated absorption cross section for  $^{149}\text{Sm}$  taken from Evaluated Nuclear Data File -ENDF-B-VI-8 ([14]) in Figure 1. We can see that the low energy part of cross section is described with (2) rather well.

At first we investigated the influence of the uranium content in the core, the amount of water in it and the core temperature on the effective absorption cross section. We took the core composition in accordance with our previous work [12]. The Uranium weight percent in the dry ore at the beginning of reactor operation was taken to be 38.4, 49.4 and 59.6%. This values covered the uranium samples composition of the Zone 2 that we investigated in this work. For example, the uranium content of the samples for borehole SC36 of Zone 2(as described by Ruffenach [15]) are presented in Table I.

TABLE I:

Sample	U content, %	$^{235}\text{U}/^{238}\text{U}$
SC36-1408/4	23.3	0.00545
SC36-1410/3	57.8	0.00527
SC36-1413/3	37.2	0.0041
SC36-1418	57.9	0.00574

The core water content in our calculations changed from 0.355 volume% to 0.455 volume%. We took the pressure of the core during its operation to be 100 MPa. The composition of other ore elements was given in [12]. This composition is given in Table II for the dry ore in comparison with the composition given in [13]. The difference between two compositions is rather small and does not influence seriously the calculated effective cross section.

We present in Figure 2 the effective cross section of  $^{149}\text{Sm}$  calculated by code MCNP-4C [16] for different temperatures of the fresh core and for different content of uranium and water. The experimental ( $1\sigma$ ) bounds on the measured cross section in different samples obtained in the work [9] are shown in the same Figure. The calculation was performed with the Monte Carlo method for the neutron spectrum which is set in the small cell. Locally, the neutron spectrum in the reactor can be approximated in the same manner. Also, these results are presented in Table III.

The effective cross section is lowered with the increase of uranium content in the core and with decreasing of the water content. The temperature dependence of the cross section has the maximum. (In the calculation the dependence of the water density on temperature [12] was taken into account). The position of the maximum is changed with the increasing of the uranium amount. Experimental boundaries of Damour and Dyson with  $2\sigma$  errors include all the calculated values. It shows that the hypothesis about the constancy of effective absorption cross section of  $^{149}\text{Sm}$

during the past 2 billion years is valid with the equal precision. Nevertheless, this analysis was done for the fresh core and the influence of fuel burn-up on the value of the effective cross section needs further investigation. This will be analyzed in the next section.

### III. DEPENDENCE OF THE EFFECTIVE CROSS SECTION ON BURN-UP

During the reactor operation, the fuel products are accumulated in the reactor core. Some products intensively absorb neutrons due to its high absorption cross section. This causes the modification of the reactor spectrum as far as the reactor burnt-up. To investigate this issue, we performed the realistic calculations of burn-up with the help of reactor code MCU-REA [17]. The parameters of the reactor was taken to be closed to one in Zone 2: the duration of reactor operation was about 800 thousand years, the accepted reactor power - 25 kW ( $1.22 \cdot 10^{-4}$  W/cm<sup>3</sup>) [12]. Kinetics of the concentrations of actinides and fuel products were calculated according to the following system of equations

$$\frac{dA_n}{dt} = -(\lambda_n + \hat{\sigma}_n \hat{\phi})A_n + \sum_i \lambda_{i,n} A_i. \quad (3)$$

Where  $A_n$  - concentration on nuclide number  $n$ ,  $\hat{\phi}$  - effective neutron flux,  $\hat{\sigma}_n$  - effective absorption cross section of  $n$ -th nuclide,  $\lambda_n$  - its decay constant,  $\lambda_{i,n}$  - decay constant of nuclide number  $i$  decay of which lead to nuclide number  $n$ . Physical values of  $\hat{\sigma}_n$  and  $\hat{\phi}$  were calculated in the nodes of the time grid by the Monte Carlo method. In the calculation all complicated physics of nuclear reactor theory was taken into account. That is the resonance structure of cross sections of fuel and constituent nuclides, the real reactor neutron spectrum established at the time  $t$  of the reactor operation, the accumulation of fission products and transuranium elements etc. The calculated dependence of the effective cross section  $\hat{\sigma}_{Sm149}$  on neutron fluence  $\tau$

$$\tau = \int \hat{\phi}(t) dt$$

is shown at Figure 3. In this calculation the uranium content in the core was 49.4% at the beginning of the reactor operation, and the amount of water was  $\omega_{H_2O} = 0.455$ . The temperature of the core was taken to be 500K. At the beginning of the reactor operation the effective cross section of  $^{149}Sm$  was 80.94 kb and at the end of operation ( $\tau \approx 1.0 \text{ kb}^{-1}$ ) it changed to 81.99 kb. The change is about 1.3%. For different core content and different temperatures the situation is nearly the same - the effective cross section of  $^{149}Sm$  is nearly constant during the reactor operation. Thus, the core composition and the temperature of the core has the main effect on the value of the effective absorption cross section of the Samarium. The spectrum modification uncertainties due to fuel burn-up have a minor importance.

### IV. SAMPLE SC52-1472

Earlier it was shown that the main uncertainty in determination of the  $^{149}Sm$  effective absorption cross section contributes the sample variation of the uranium amount, water content and the core temperature. The main idea of the following investigation is to restrict considerably and locally the conditions of the chain reaction occurring. For this purpose we considered the burn-up of sample S52-1472 from the reactor Zone 3 [19]. This approach is similar to one adopted in [13]. In this paper, the burn-up of the samples from the Zone 2 and Zone 10 were analyzed. The main difference of our approach from one in [13] is that we fix the amount of water in the sample during the reaction

TABLE II: Present day composition of the empty rock

Chemical composition	% by weight (this work)	% by weight (from [13])
SiO <sub>2</sub>	43.00	42.14
Al <sub>2</sub> O <sub>3</sub>	25.73	27.93
FeO, Fe <sub>2</sub> O <sub>3</sub>	19.0	17.49
MgO	10.43	9.04
MnO	-	1.06
K <sub>2</sub> O	1.84	2.35

operation and the temperature of the neutron spectrum. Also, the Samarium amount in the sample was determined before the beginning of the chain reaction.

For the particular sample we definitely know the content of the uranium in it and initial composition of the rear earth impurities. We know the burn-up of the uranium in the sample at the end of the reactor operation. The duration of the chain reaction can be determined if the contribution of the  $^{238}\text{U}$  ( $\alpha$ ) and  $^{239}\text{Pu}$  ( $\beta$ ) nucleus to fission is known. This contribution can be determined from the following nuclear isotope ratios in the sample:

$$\begin{aligned} &^{150}\text{Nd}/(^{143}\text{Nd}+^{144}\text{Nd}+^{145}\text{Nd}+^{146}\text{Nd}), \\ &^{154}\text{Sm}/(^{147}\text{Sm}+^{148}\text{Sm}), \\ &(^{157}\text{Gd}+^{158}\text{Gd})/(^{155}\text{Gd}+^{156}\text{Gd}). \end{aligned}$$

It follows from the fact that the yield of isotopes  $^{150}\text{Nd}$ ,  $^{154}\text{Sm}$  and  $^{157}\text{Gd}$ ,  $^{158}\text{Gd}$  in fission strongly depends on the nuclei that undergoes fission. The calculation shows that for a particular sample about 2% of fissions were due to  $^{238}\text{U}$  and  $^{239}\text{Pu}$ .

The production of the  $^{239}\text{Pu}$  nuclei in the reaction of neutron absorption by the  $^{238}\text{U}$  nuclei also influences the reactor energy balance. The Plutonium isotope have the half life equal to 24110 years and transmitted into the  $^{235}\text{U}$  isotope after the  $\alpha$ -decay. We identify the conversion factor as  $C$  [9]. The duration of reactor operation can be evaluated then as follows:

$$\Delta t = \frac{\hat{\sigma}_{239,f}\hat{\sigma}_{235}}{\hat{\sigma}_{235,f}} \frac{1 - \alpha - \beta}{\beta} \frac{C\tau}{\lambda_{239}}. \quad (4)$$

In this equation  $\hat{\sigma}_{235,f}$ ,  $\hat{\sigma}_{239,f}$  – is the effective fission cross section of  $^{235}\text{U}$  and  $^{239}\text{Pu}$  respectively,  $\hat{\sigma}_{235}$  – absorption cross section of  $^{235}\text{U}$ ,  $\lambda_{239}$  – decay constant of  $^{239}\text{Pu}$  nuclei. Based on the produced energy in a volume unit and the duration of the nuclear chain reaction the mean nuclear power release in the sample can be evaluated.

The main factor that influences the spectrum index is the amount of water in the sample [13]. The very precise measure of the spectrum index is the value of the  $^{147}\text{Sm}$  absorption cross section. The approximate relation between

TABLE III: Effective neutron cross section of  $^{149}\text{Sm}$

	$\hat{\sigma}_{149\text{Sm}}$	
	38.4 wt.% U in ore	
	$\omega_{H_2O} = 0.355$	$\omega_{H_2O} = 0.455$
300	68.2	68.3
400	75.5	76.6
500	78.9	80.8
600	79.0	81.3
700	76.4	79.0
800	72.1	75.1
	49.4 wt.% U in ore	
	$\omega_{H_2O} = 0.355$	$\omega_{H_2O} = 0.455$
300	67.7	68.1
400	73.3	74.9
500	75.5	78.2
600	74.9	77.8
700	71.4	74.8
800	65.7	69.8
	59.6 wt.% U in ore	
	$\omega_{H_2O} = 0.355$	$\omega_{H_2O} = 0.455$
300	66.4	67.4
400	70.5	72.8
500	71.7	74.8
600	70.0	73.8
700	65.7	70.0
800	59.0	63.9

the effective absorption cross section of the  $^{147}\text{Sm}$  and the spectrum index  $r$  as follows:

$$\hat{\sigma}_{Sm147} = 0.88 r,$$

where cross section is in kilo barns. The absorption cross section of the  $^{147}\text{Sm}$  isotope, on the other hand, influences the isotopic ratios of Sm in the sample, notably the content of  $^{148}\text{Sm}$ , because this isotope is not produced within the fuel fission. Varying the amount of water in the sample we can reproduce the experimental ratio for the  $^{148}\text{Sm}$  isotope. Thus, the procedure fixes the spectrum index.

The next unknown quantity is the core temperature. The neutron spectrum in reactor significantly depends on the core temperature. The higher temperatures, the more probable higher neutron energies. In Figure 4 we present two neutron spectrums calculated according to code MCNP-4C for  $T = 400$  and  $600$  K as compared to the effective absorption cross section by  $^{149}\text{Sm}$ . To fix the temperature we used the experimental ratio of two Lu isotopes in the sample:  $^{176}\text{Lu}/^{175}\text{Lu}$ . As for a  $^{149}\text{Sm}$  isotope, the  $^{176}\text{Lu}$  isotope has a low energy resonance. The energy of the resonance is 141.3 meV. The absorption cross section is plotted in Figure 5 in comparison with two neutron spectrums with  $T = 400$  and  $600$  K. As you can see from the comparison, the neutron spectrum shifted to the right, when its temperature increased considerably overlapping the resonance cross section for higher temperature. Determining the spectrum temperature from the description of the Lu isotope ratio in the sample and the spectrum index from the description of the  $^{148}\text{Sm}$  isotope ratio we mainly determined the form of the neutron spectrum in the equation (1). Using the  $^{149}\text{Sm}$  cross section from (2) we can determine the *theoretical effective cross section* of this isotope.

The main parameters of the sample and the time parameters of the reactor in Zone 3 are presented in Table IV [19, 20]. The nuclide composition of the sample (adopted in our calculations) at the beginning of the burn-up are presented in Table V. The density of the dry ore was determined from content of the Uranium in it (see [12]). It was equal to  $3.465 \text{ g/cm}^3$ . The isotope ratio of Uranium at the beginning of the reactor operation was determined using the age of the reactor and the current normal isotope ratio for Uranium. At the beginning of the reactor life this ratio was equal to:  $^{235}\text{U}/^{238}\text{U} = 3.6\%$ . The composition of the empty rock was calculated in accordance with the Table 2. The amount of Samarium in the sample before operation was determined based on its current concentration using the fact that  $^{144}\text{Sm}$  isotope is not produced in fission but occurred in the sample before the beginning of the chain reaction. The other isotopes were distributed according to normal isotopic ratio. The same procedure was applied to Neodymium but based on the concentration of the  $^{142}\text{Nd}$  isotope. We neglected the chemical reshuffling after the chain reaction ended.

TABLE IV:

U content in sample, wt.%	Fluence, $\text{kb}^{-1}$	Duration, years	Age, MY	Specific power, $\text{W/cm}^3$
$0.32 \pm 0.01$	0.228	300 000	1.93	$0.45 \cdot 10^{-4}$

The value of the *experimental cross section* for  $^{149}\text{Sm}$  can be determined from the description of the  $^{149}\text{Sm}$  isotope concentration in the sample. As the sample nuclide composition was fixed, we could change only the temperature of the core to vary the concentration of this isotope at the end of the burn-up. We have made three computations of the sample burn-up at three different temperatures: 300, 400 and 500 K. We used the burn-up utility MURE [21] which is based on MCNP code. The isotope ratios of the Sm at the end of the burn-up is presented in Table VI in comparison with the experimental data. Table VI confirms that the temperature of the core should be equal to 367 K (using linear

TABLE V: Initial density of SC56-1472 sample

Element	Concentration, atoms/barn cm	Element	Concentration, atoms/barn cm	Element	Concentration, atoms/barn cm
U-235	$0.10751 \cdot 10^{-3}$	Nd-142	$0.20149 \cdot 10^{-6}$	Sm-144	$3.186 \cdot 10^{-9}$
U-238	$0.29898 \cdot 10^{-2}$	Nd-143	$0.90376 \cdot 10^{-7}$	Sm-147	$0.14839 \cdot 10^{-7}$
O	0.058581	Nd-144	$0.17631 \cdot 10^{-6}$	Sm-148	$0.11126 \cdot 10^{-7}$
H	0.046799	Nd-145	$0.61485 \cdot 10^{-7}$	Sm-149	$0.13680 \cdot 10^{-7}$
Si	$0.75364 \cdot 10^{-2}$	Nd-146	$0.12742 \cdot 10^{-6}$	Sm-150	$0.73055 \cdot 10^{-8}$
Al	$0.53157 \cdot 10^{-2}$	Nd-148	$0.42225 \cdot 10^{-7}$	Sm-152	$0.26480 \cdot 10^{-7}$
Mg	$0.27296 \cdot 10^{-2}$	Nd-150	$0.41484 \cdot 10^{-7}$	Sm-154	$0.22520 \cdot 10^{-7}$
K	$0.41208 \cdot 10^{-3}$				
Fe	$0.27191 \cdot 10^{-2}$				

fit) to describe the experimental concentration of the  $^{149}\text{Sm}$  in the sample. In Table VII effective cross section of  $^{149}\text{Sm}$  for this temperatures is presented. From this table it is followed that the cross section of  $^{149}\text{Sm}$  at 367 K is equal to 62.91 kb. Thus, the value of the experimental effective absorption cross section we determined is equal to 62.91 kb. This value can be compared with the value obtained for considered sample S56-1472 in [9]. In this work for this cross section the value 72 kbarn was received. Thus, our experimental cross section of  $^{149}\text{Sm}$  is considerably lower than in [9]. This disagreement can be attributed to the procedure used in [9] to obtain an experimental cross section. In this analysis the contribution of  $^{238}\text{U}$  and  $^{239}\text{Pu}$  to fission was not taken into account. The influence of this nuclei could be rather strong, and, if we take them into account, the experimental cross section would be considerably smaller.

In Table VIII, the calculated isotope ratios for Neodymium nuclei in the sample at the end of burn-up are compared to the experimental data. The agreement is rather good. Particularly, the experimental isotope ratio for the  $^{150}\text{Nd}$  isotope is reproduced in our calculations. As you can see from the table, the temperature dependence of isotopic ratios for Neodymium is weak.

TABLE VI: Calculated isotopic ratios of Sm at the end of burn-up

Sm isotope	Temperature, K			Exp. distrib.
	300	400	500	
144	0.53	0.54	0.53	$0.53 \pm 0.005$
147	50.93	51.29	50.99	$51.55 \pm 0.06$
148	2.70	2.74	2.71	$2.73 \pm 0.01$
149	1.47	1.25	1.19	$1.32 \pm 0.005$
150	25.60	25.58	25.93	$25.36 \pm 0.03$
152	13.31	13.10	13.16	$12.94 \pm 0.01$
154	5.46	5.51	5.48	$5.57 \pm 0.01$

TABLE VII: Effective cross section of  $^{149}\text{Sm}$  and  $^{175,176}\text{Lu}$  for different temperatures

Isotope	Temperature, K		
	300	400	500
Sm149	55.67	66.47	72.49
Lu175	0.0841	0.0869	0.0845
Lu176	4.198	5.232	6.299

TABLE VIII: Calculated isotopic ratios of Nd at the end of burn-up

Nd isotope	Temperature, K			Exp. distrib.
	300	400	500	
142	5.83	5.84	5.84	$5.75 \pm 0.02$
143	24.17	24.17	24.16	$24.33 \pm 0.04$
144	26.85	26.77	26.85	$26.84 \pm 0.04$
145	16.57	16.62	16.57	$16.57 \pm 0.03$
146	15.23	15.21	15.23	$15.15 \pm 0.03$
148	7.61	7.66	7.62	$7.62 \pm 0.02$
150	3.74	3.73	3.74	$3.73 \pm 0.01$

Independently we can determine the temperature of the core during the reactor operation from the lutetium isotope ratio in the sample. Effective cross sections of lutetium isotopes at different temperatures are presented in Table VII. At the beginning of the reactor operation we took the lutetium isotope ratio to be normal:

$$^{176}\text{Lu}/^{175}\text{Lu} = 0.0266.$$

The absorption of neutron by the  $^{175}\text{Lu}$  isotope leads to the isotope  $^{176}\text{Lu}$ . Part of the  $^{176}\text{Lu}$  nuclei in this reaction is produced in the metastable state. The spin of this metastable state considerably differ from the spin of the ground state. Due to this fact, the radiation transition from the metastable state to ground one is forbidden and the main mode of decay of the metastable state is the  $\beta^-$  decay to  $^{176}\text{Hf}$  nuclei. The total experimental absorption cross section

of the thermal neutron by  $^{175}\text{Lu}$  is equal to  $23.3 \pm 1.1$  b [22]. The experimental cross section of production of  $^{176}\text{Lu}$  in the ground state is equal to  $6.6 \pm 1.3$  b [22]. So the experimental yield of the ground state in the neutron absorption by  $^{175}\text{Lu}$  nuclei is equal to  $0.283 \pm 0.056$  with  $1\sigma$  error. The yield of lutetium isotopes in fission is negligibly small. The  $^{175}\text{Lu}$  nuclei can be produced only in the reaction of neutron absorption with  $^{174}\text{Yb}$  nuclei. The effective absorption cross section of neutron by  $^{176}\text{Lu}$  nuclei is presented in Table VII. It increases within the temperature increasing. At higher temperatures  $^{176}\text{Lu}$  nuclear burns up better in the sample. So the ratio  $^{176}\text{Lu}/^{175}\text{Lu}$  in the sample can help to determine the spectrum temperature. The dependence of this ratio at the end of burn-up cycle on the core temperature is presented in Figure 6. In good approximation this dependence is linear. Experimentally this ratio is equal to  $^{176}\text{Lu}/^{175}\text{Lu} = 0.0107 \pm 0.0001$  [19]. In Figure 6 we can determine what was the core temperature during the reactor operation. It's equal to 455 K. The cross section of the  $^{149}\text{Sm}$  at this temperature amounts to 69.78 kb from Table 6. The disagreement between the  $^{149}\text{Sm}$  experimental effective cross section obtained from the concentration of this isotope in the sample and the theoretical value of this cross section calculated with the help of the evaluated neutron spectrum is equal to 6.9 kb.

Such disagreement in the  $^{149}\text{Sm}$  cross section experimental and theoretical values can be explained with the help of the hypotheses that the shift of the position of the  $^{149}\text{Sm}$  low laying resonance occurred for about the past  $2 \cdot 10^9$  years time. The dependence of the  $^{149}\text{Sm}$  cross section on the shift of the resonance  $\Delta E_r$  is shown in Figure 7. This dependence was calculated for the core temperature equal to 400 K at the end of the burn-up for the real neutron spectrum. As you can see in the Figure, the theoretical value of the  $^{149}\text{Sm}$  cross section equal to 69.78 kb can be explained by the shift of the resonance position by amount  $\Delta E_r = -7.6$  meV. However, we must take into account uncertainties in the temperature determined with the help of lutetium. This analysis is performed in the next section.

## V. ANALYSIS OF THE AMBIGUITIES

The lutetium isotope ratio at the end of burn up is sensitive to the ground state  $^{176}\text{Lu}$  yield in the absorption of neutron by  $^{175}\text{Lu}$  nuclei. This yield have rather high uncertainty:  $0.283 \pm 0.056$  with  $1\sigma$  error. We have investigated the dependence of the  $^{176}\text{Lu}/^{175}\text{Lu}$  ratio at the end of burn-up on this yield. For the yield  $0.283 - 3 \cdot 0.056$  we obtained the temperature equal to 364 K. On the other side, for the yield  $0.283 + 3 \cdot 0.056$  the temperature is to be equal to 525 K. As a result the core temperature determined from the lutetium isotope ratio is in the range

$$364 \text{ K} < T < 525 \text{ K}$$

at 99% C.L. The values of the  $^{149}\text{Sm}$  cross section (as it follows from Table 7) are in the range 62.6 kb to 74.0 kb. Following the dependence of the  $^{149}\text{Sm}$  cross section on a resonance shift (see Figure 7) we conclude that the resonance shift should be in the range

$$-11.3 \text{ meV} < \Delta E_r < 0.8 \text{ meV},$$

to satisfy the experimental condition. So the zero resonance shift is not excluded.

## VI. DISCUSSION OF THE RESULTS

Constraints for the shift of the  $^{149}\text{Sm}$  resonance obtained in the previous section can be converted into the limits for the variation of the fine structure constant  $\alpha$  (see [12])

$$\frac{\Delta\alpha}{\alpha} = \frac{\Delta E_r}{M} \quad (5)$$

where:  $M$  is estimated in [9] as  $-(1.1 \pm 0.1)$  MeV. Using this value of  $M$  we obtain constraints for the variation of the fine structure constant

$$-0.7 \cdot 10^{-9} \leq \frac{\Delta\alpha}{\alpha} \leq 1.0 \cdot 10^{-8} \quad (6)$$

during the past  $2 \cdot 10^9$  years. Dividing the equation (6) by the age of the reactor  $1.93 \cdot 10^9$  years we get the following limit (99% C.L.) for the time derivative of  $\alpha$

$$-0.35 \cdot 10^{-18} \text{ yr}^{-1} \leq \frac{\Delta\dot{\alpha}}{\alpha} \leq 5 \cdot 10^{-18} \text{ yr}^{-1}. \quad (7)$$

This constraint is of the next higher order than in our previous work [12].

The only evidences obtained till now about the variation of fundamental constants include astrophysical observations of the variation of the fine structure constant for  $0.2 \leq z \leq 4.2$  and the change of the electron to proton mass ratio  $\mu$  for  $z \sim 2.8$  [23–25]. If this variations are due to fundamental scalar field, it causes also the violation of the Weak Equivalence Principle (WEP) of General Relativity [26–29]. The reported values of  $\alpha$  and  $\mu$  variation in astrophysical observations, if they are true, predict the violation of WEP on the observable level in future experiments.

Supersymmetric scenarios of the unifications of forces naturally lead to the fundamental cosmon scalar field coupling with the matter and electromagnetic energy. The variation of scalar field over the cosmological times in the unified scenarios leads to the variation of nuclear couplings. Consistency of different scenarios of unification with the reported variations of the fundamental constants and with the constraints for the violation of WEP was analyzed in [7, 27, 29, 30]. In this analysis the Oklo's constrain for the variation of  $\alpha$  serves as an additional rigid boundary condition to the evolution of the cosmon field and generally provides the severely restriction for the possible unification couplings and unification models.

In this paper we take into account only the influence of coulomb energy on the position of resonances. It is highly unrealistic that the modification of fine structure constant is accompanied with changing of strong coupling constant in such a manner that the total nuclear energy is unchanged. Contrary it can be expected that the constraints will be ever harder [5], if we admit the unification forces hypotheses.

As a result, we state that the variation of  $\alpha$  during the past 2 billion years is not larger than  $1.0 \cdot 10^{-8}$ . For this result confirmation additional investigation of burn-up of the reactor Oklo other samples is necessary

## VII. ACKNOWLEDGMENTS

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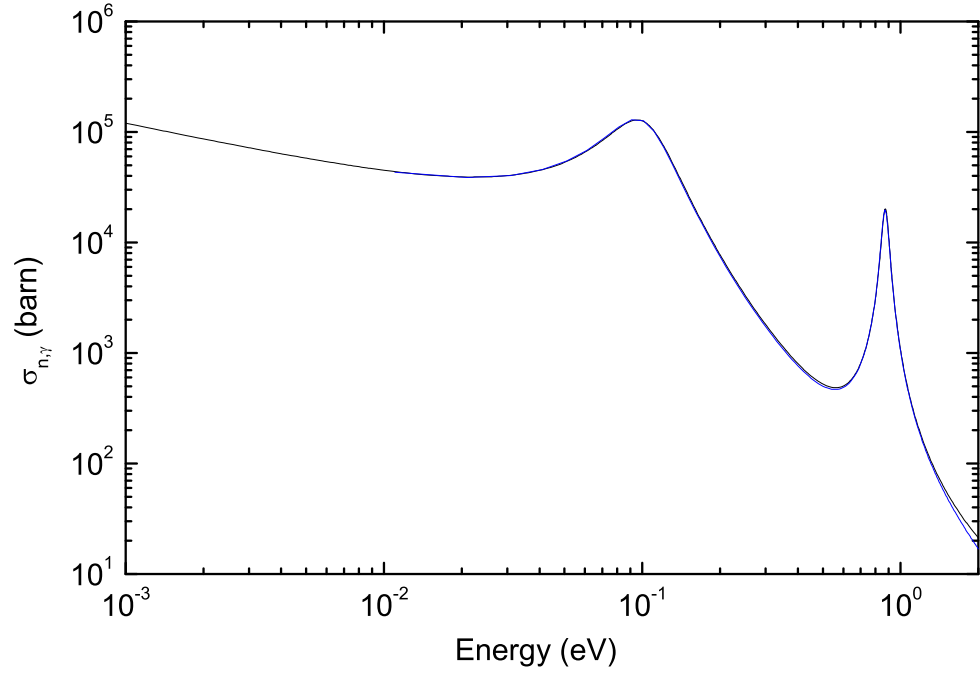


FIG. 1: ENDF capture cross section of  $^{149}\text{Sm}$  - black solid line. Two resonance approximation of capture cross section - blue solid line.

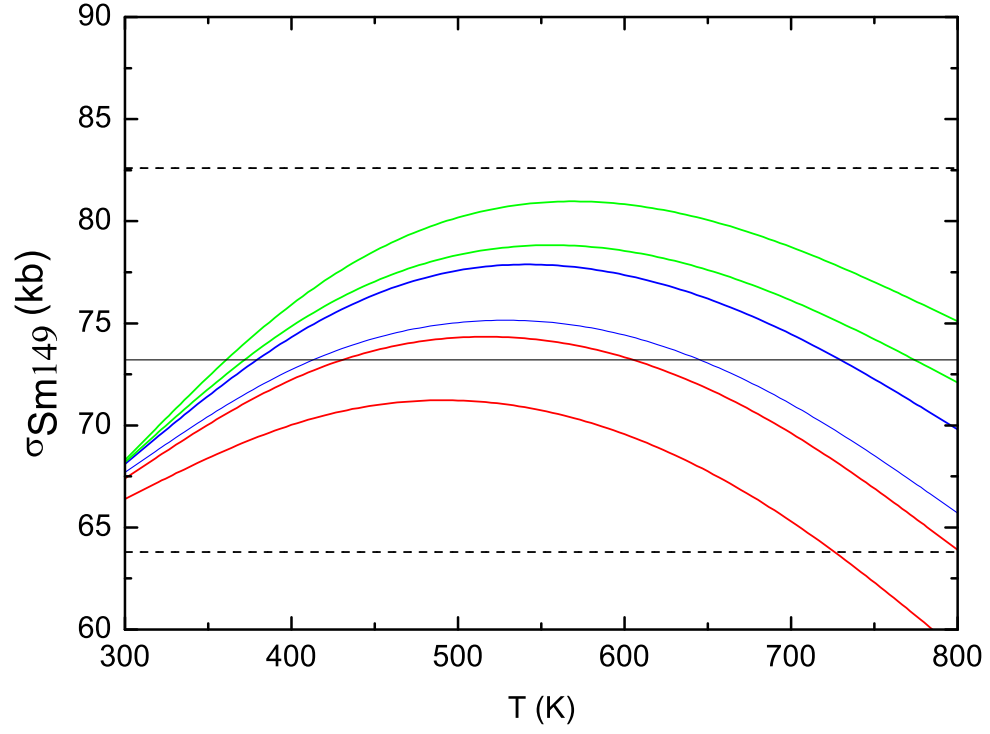


FIG. 2: Dependence of effective capture cross section of  $^{149}\text{Sm}$  on the core temperature. Green lines - uranium content 35%, upper curve -  $\omega_{H_2O}=0.355$ , lower curve -  $\omega_{H_2O}=0.455$ . Blue lines - uranium content 45%, upper curve -  $\omega_{H_2O}=0.355$ , lower curve -  $\omega_{H_2O}=0.455$ . Red lines - uranium content 55%, upper curve -  $\omega_{H_2O}=0.355$ , lower curve -  $\omega_{H_2O}=0.455$ . Also, the error corridor of  $\sigma_{Sm149}$  measured values from work [9]:  $73.2 \pm 9.4$  kb is shown.

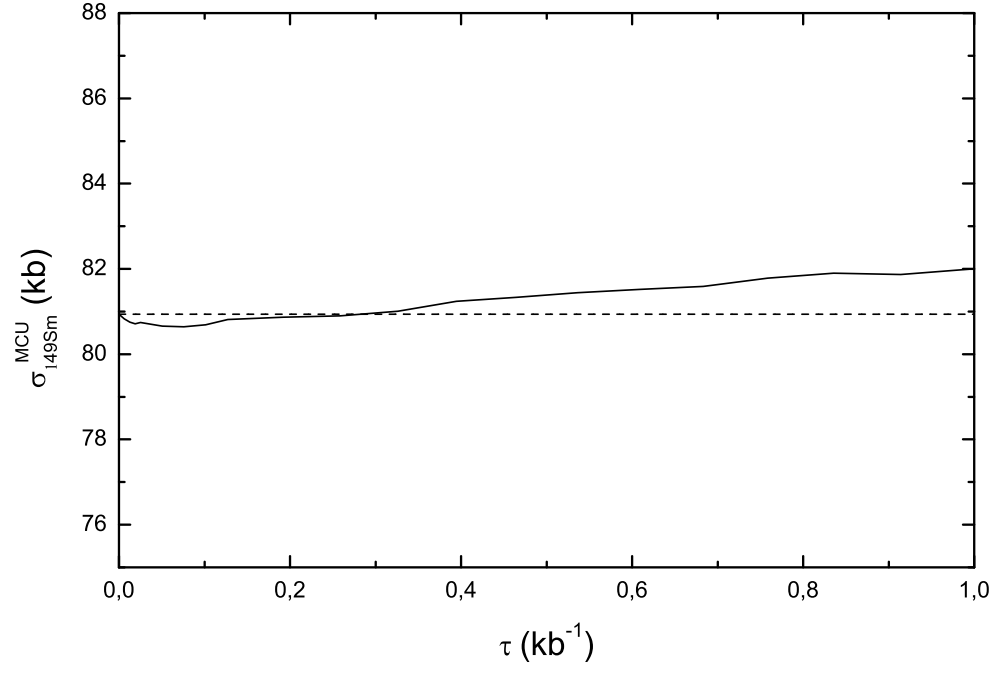


FIG. 3: Dependence of the calculated effective capture cross section of  $^{149}\text{Sm}$  on  $\tau$ .

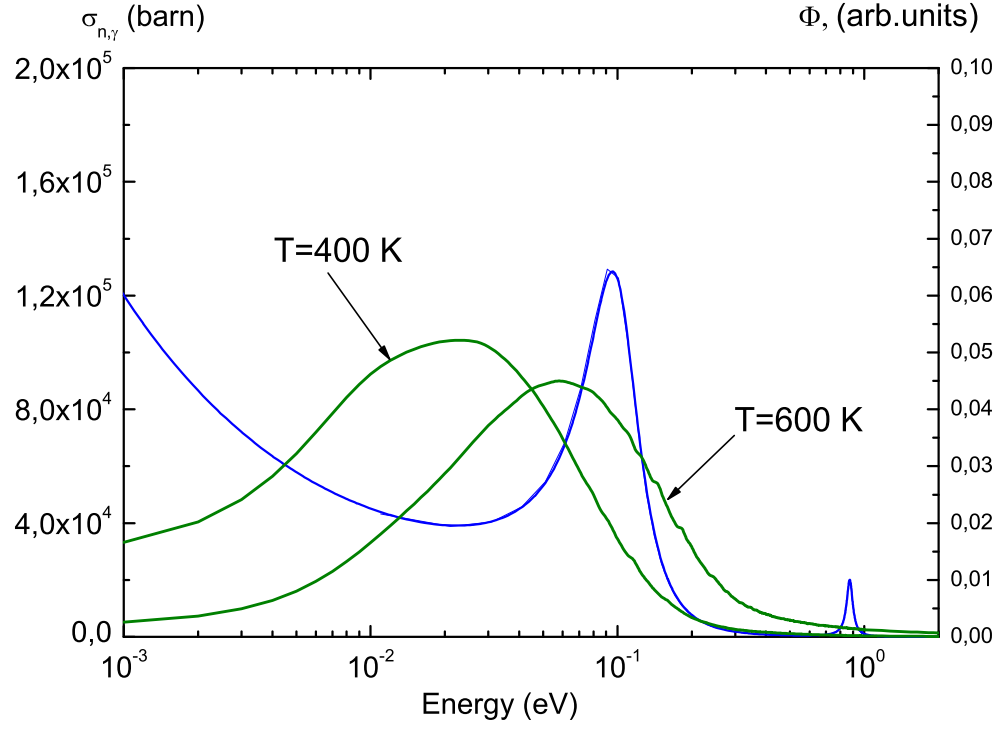


FIG. 4: Energy dependence of the effective absorption cross section of the  $^{149}\text{Sm}$  (left, blue line). Calculated neutron fluxes for temperatures 400 and 600 K (right, green lines).

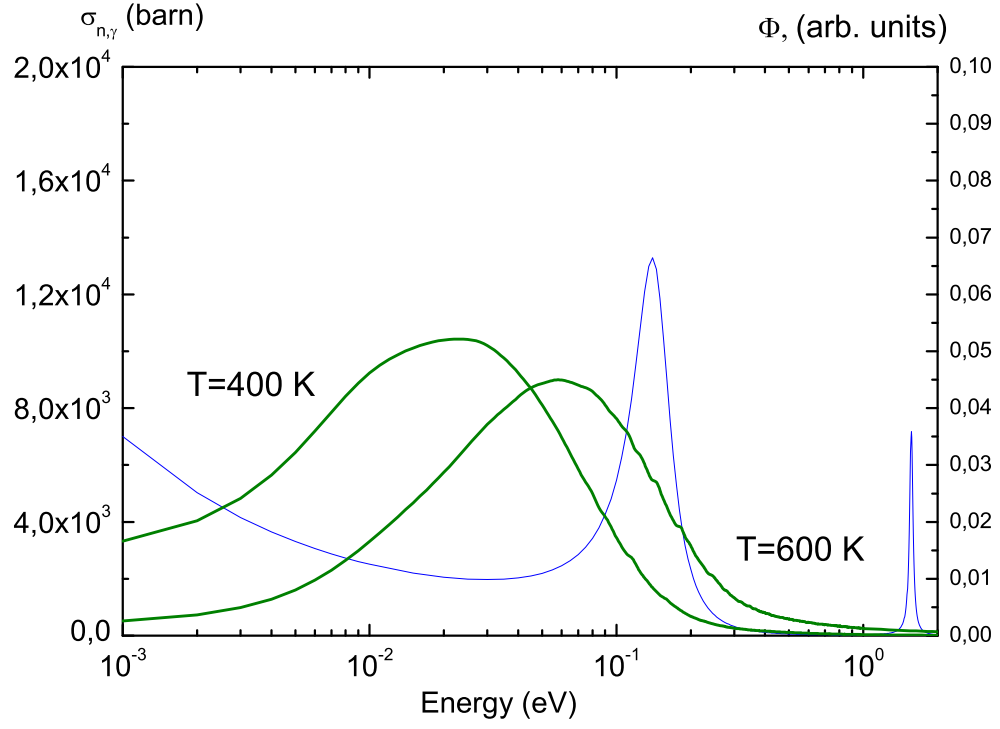


FIG. 5: Energy dependence of the effective absorption cross section of the  $^{176}\text{Lu}$  (left, blue line). Calculated neutron fluxes for temperatures 400 and 600 K (right, green lines).

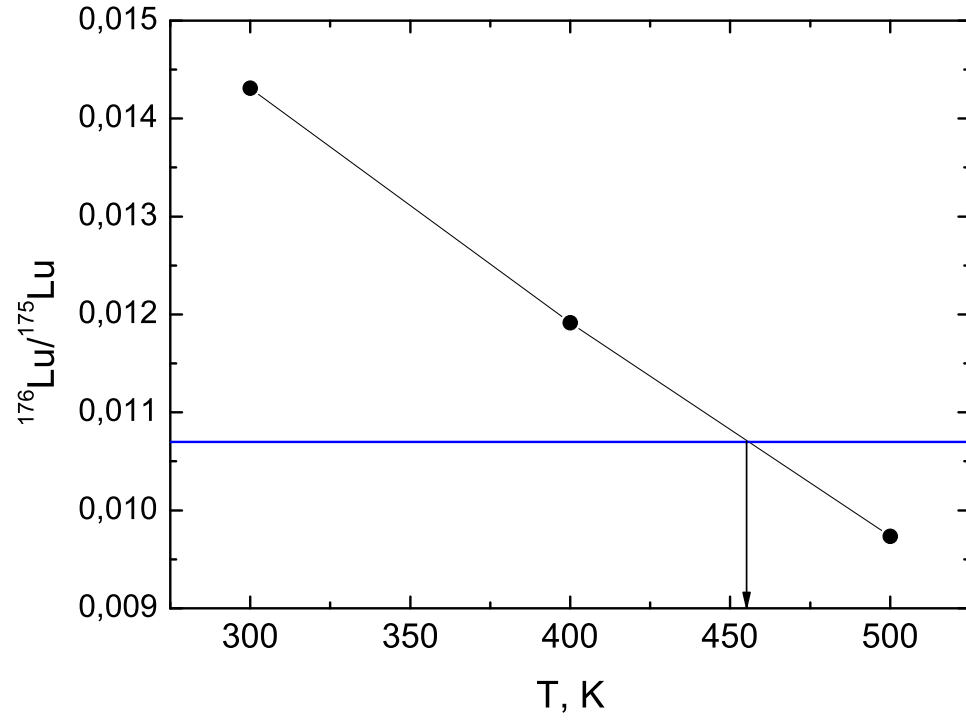


FIG. 6: Dependence of  $^{176}\text{Lu}/^{175}\text{Lu}$  ratio at the end of burn up cycle on the temperature.

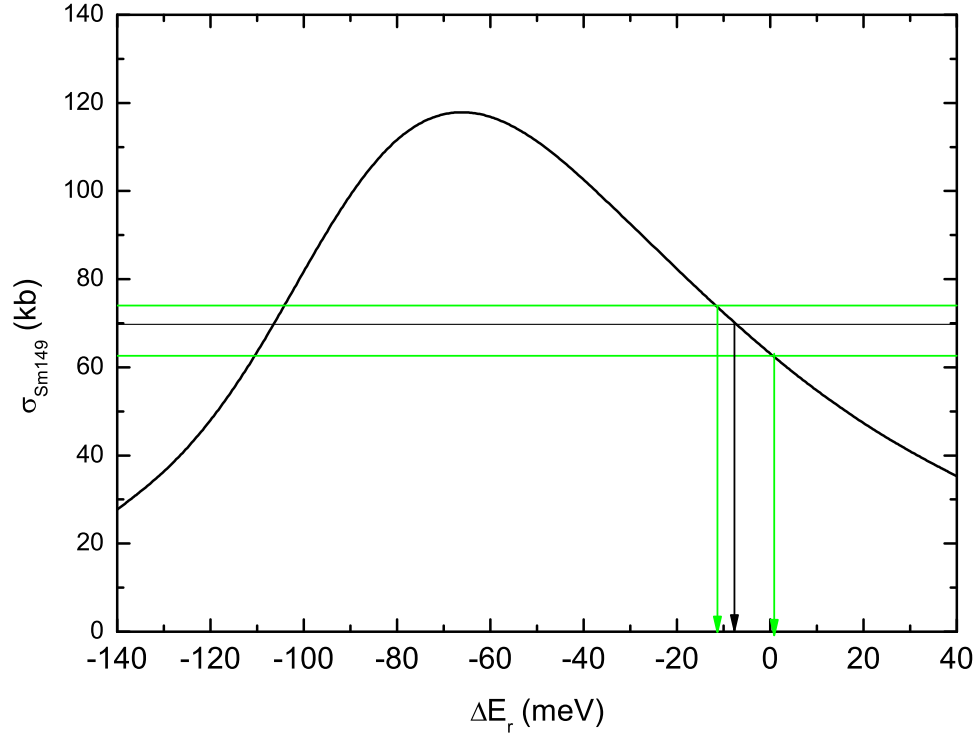


FIG. 7: Dependence of the  $^{149}\text{Sm}$  cross section on the resonance shift.